Dynamical Diffraction Effects and the Ultrafast Modulation of X-rays

David A. Reis^{1,2}, Matt F. DeCamp¹, Philip H. Bucksbaum^{1,2}, John M. Caraher¹, Roy Clarke¹, Charlie W. S. Conover^{2,3}, Eric Dufresne¹, Roberto Merlin¹, Vladimir A. Stoica¹, Jared Wahlstrand¹, and Bernhard Adams⁴

¹University of Michigan, Department of Physics ²University of Michigan, Center for Ultrafast Optical Science ³Colby College ⁴Argonne National Lab

Introduction

Using an ultrafast laser to produce picosecond pulses of coherent acoustic phonons, we are able to interrupt the anomalous transmission of x-rays (Borrmann Effect). In addition to the loss of anomalous transmission, oscillations in the diffracted intensity are induced on a time scale that corresponds to Pendellösung oscillations from a thin crystal expanding at the speed of sound. The use of the Laue geometry allows study strain propagation throughout the entire crystal bulk. Dynamic effects in x-ray diffraction may lead to the development of a femtosecond x-ray switch that will enable researchers to probe atomic positions during chemical reactions and phase transitions with unparalleled temporal resolution.

Methods and Materials

The experiments use a 50 fs Ti:sapphire laser operating at 1 kHz to impulsively strain the crystal through ultrafast heating. The laser is phaselocked to the accelerator rf with coarse timing between the laser and x-rays controllable to 19 ps. The data is taken in the usual pump-probe manner. The diffracted ('H') and forward diffracted ('O') beams of a single x-ray pulse are measured simultaneously with avalanche photodiodes as a function of time delay between the laser and the x-rays. The undulator is tuned for 10 keV x-rays and a Si (111) liquid nitrogen cooled double crystal monochromator sets the bandwidth at about 1.4 eV. The sample is a 280µm thick Ge [001].

Results

FIG. 1 shows the propagation of a phonon pulse traveling between the entrance and exit faces of the Ge sample. The asymmetric reflection, (20-2), was chosen as the probe because of its sensitivity to strain perpendicular to the surface. The ability to resolve the fast edge is limited by the x-ray bunch length and timing jitter (~100ps). FIG. 2 are time-resolved rocking curves for the diffracted and forward diffracted beams upon arrival of the phonon pulse at exit face.

Discussion

We demonstrate new dynamical effects following ultrafast laser excitation in germanium. This includes the loss (and revival) of anomalous transmission of x-rays in a time <100 ps, and the generation of Pendellösung oscillations in the diffracted and transmitted (forward-diffracted) beams. These oscillations are interpreted in terms of a

multiple crystal model where the length of the crystals increases or decreases at the speed of sound as the coherent acoustic phonon pulse traverses the crystal. We note that coherent optical phonons can modulate the x-ray pulse on a femtosecond time scale.

Acknowledgements

This work was conducted at the MHATT-CAT insertion device beamline on the Advanced Photon Source and was supported in part by the U.S. Department of Energy grant DE-FG02-00ER15031. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38. Two of us (DR, CC) acknowledge support from the National Science Foundation CUOS Fellows program. One of us (RM) acknowledges support from the Air Force office of scientific research under contract F49620-001-0328 through the MURI program.

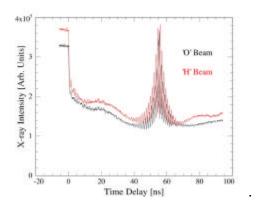


Figure 1 Time-resolved x-ray diffraction following laser induced strain Ge (20-2)

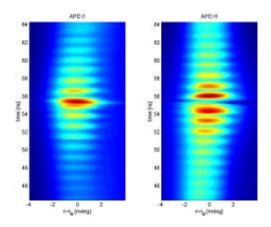


Figure 2 Time-resolved Ge (20-2) rocking curve, when strain arrives at exit face of crystal